Seasonal Variability of Trans-Pacific Transport of Carbon Monoxide (CO) in the Upper Troposphere: MLS Observations and Model Simulations

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1 Introduction

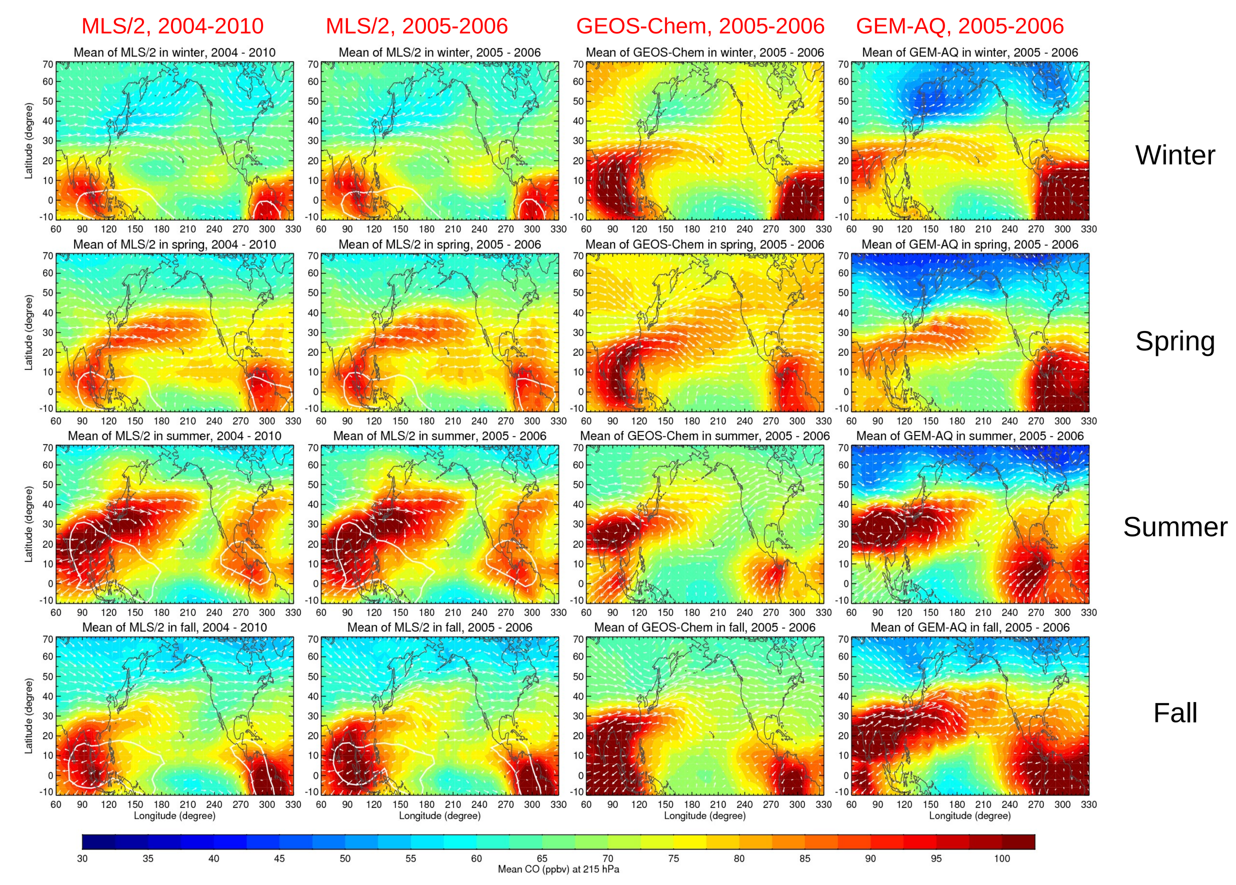
Trans-Pacific transport of air pollution has drawn much attention in this decade (e.g., Jacob et al., 2003; Heald et al., 2003; Liang et al., 2004, 2007, Wang et al., 2006; Turquety et al., 2008). Economic growth in the East Asia and South Asia is expected to increase the transport. However, most of the studies to date have focused on spring season. Liang et al. (2004) show that there is a seasonal variability in the transport of air pollutants. Recently, Jiang et al. (2007) demonstrate that trans-Pacific transport of air pollution is evident in the upper troposphere because of the convective outflow over the South Asia and East Asia (Li et al., 2005, Park et al., 2009) during Asian summer monsoon season.

In this study, we use the carbon monoxide (CO) measured by the Microwave Limb Sounder (MLS) on Aura satellite(Waters et al., 2006) at 215 hPa to characterize the seasonal variability of the trans-Pacific transport of air pollution in the upper troposphere. CO is a well established tracer of atmospheric transport and a key air pollutant. Its reaction with hydroxyl radicals (OH), which controls the atmospheric oxidation capability, is the main sink of that radical. CO abundance also affects tropospheric ozone production. MLS observations are compared with simulations from two state-of-art tropospheric air quality models GEOS-Chem (Bey et al., 2001) and GEM-AQ (Kaminski et al., 2008).

2 Seasonal means, standard deviations, occurrence probabilities of CO

Figure 1 Seasonal means of CO measured by MLS and simulated by GEOS-Chem (v8-03-01) and GEM-AQ in Northern Hemisphere winter (Dec-Jan-Feb), spring (Mar-Apr-May), summer (Jun-Jul-Aug), and fall (Sep-Oct-Nov). MLS CO (v2.2) is divided by its bias of a factor of two in this study. White contour is for 3 mg/m^3 ice water content (IWC).

• MLS observations made in year 2005 and 2006 are used to compare with model simulations. The seasonal means of MLS observations during the two yeas show little difference from the season means of observations during the six years (2004 - 2010).
• MLS observations show weak trans-Pacific transport in winter, strong transport in spring, strongest Asia outflow in summer, and relatively weak transport in fall.
• GEOS-Chem and GEM-AQ can simulate the Asia outflow and the long-rang transport of CO.
• GEOS-Chem shows the strongest transport in spring while GEM-AQ shows the strongest transport in fall.



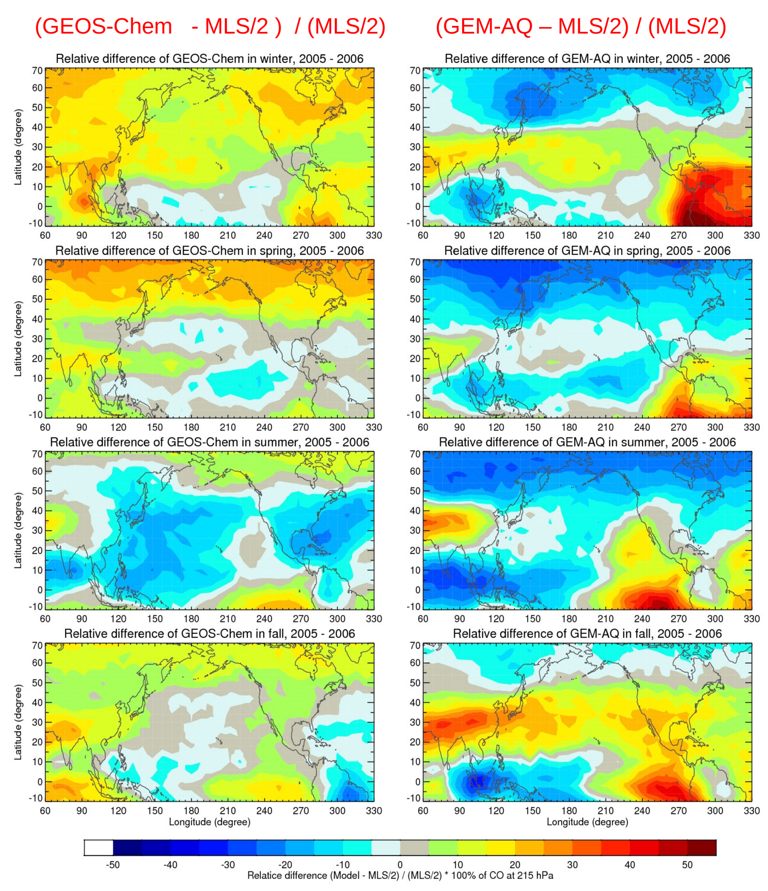
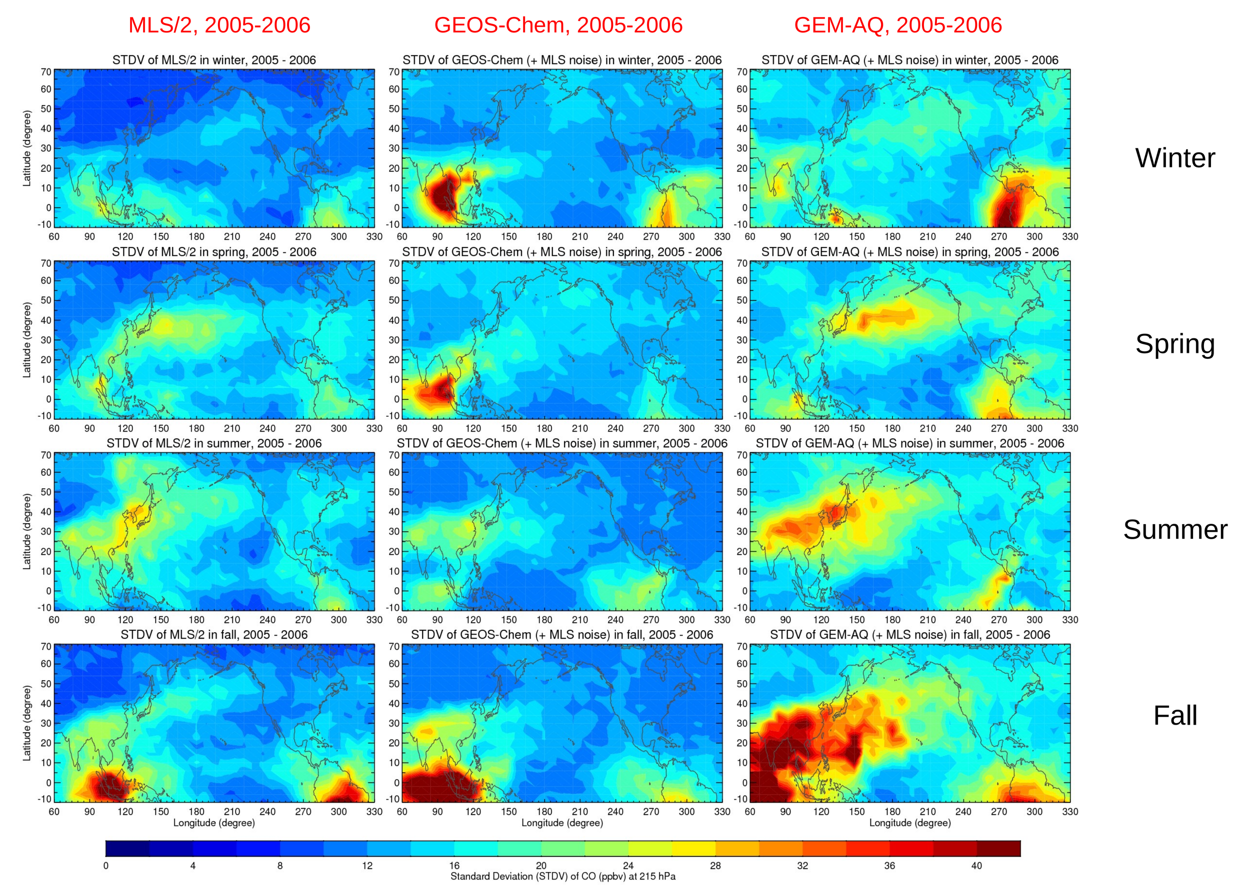


Figure 2 Relative difference of GEOS-Chem and GEM-AQ simulations compared to MLS observations in the four seasons.

• GEOS-Chem, compared with MLS, generally over-estimates CO at the northern high latitudes in all four seasons. It also over-estimates the trans-Pacific transport in winter, but slightly under-estimates the transport in spring.
• GEOS-Chem significantly under-estimates the trans-Pacific transport and the North American outflow in summer, but its results are close to MLS in fall.
• GEM-AQ, in general, under-estimates CO at the northern high latitudes in all seasons, compared with MLS. It also over-estimates CO over Asia, but under-estimates CO over Southeast Asia.
• GEM-AQ over-estimates CO over the North Pacific in winter and fall, but generally reproduces the transport-plume in spring and summer.
• GEM-AQ shows large positive bias over tropical South America.

Figure 3 Standard deviation (STDV) of MLS CO measurements and GEOS-Chem and GEM-AQ simulations in the four seasons. MLS noise is added to the model results when the standard deviations are calculated.

• GEOS-Chem, compared with MLS, has larger CO variabilities in Southeast Asia in winter, spring and fall, but smaller variabilities over the North Pacific in spring and summer.
• GEM-AQ has generally larger variabilities over Asia, and the North Pacific as well as tropical South America than MLS.



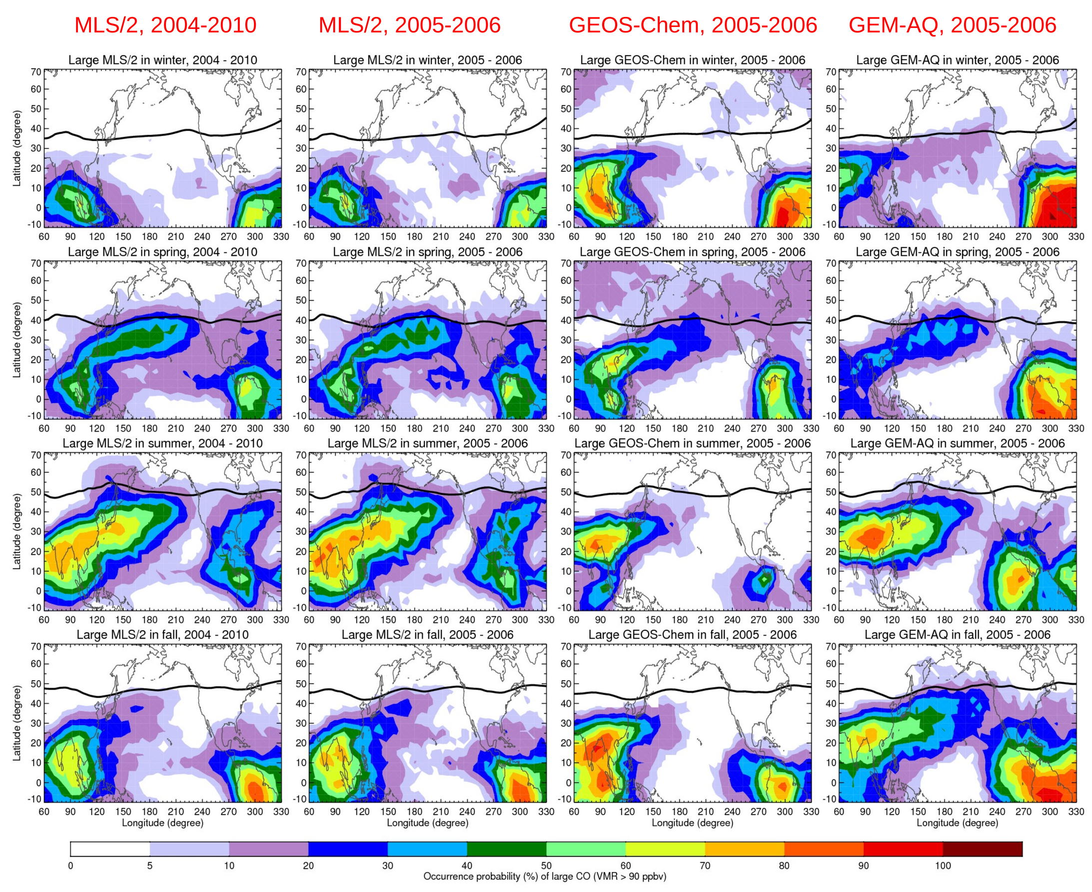


Figure 4 Occurrence probabilities of large CO abundance (> 90 ppbv) in MLS measurements and GEOS-Chem and GEM-AQ simulations, demonstrating the probabilities of transport events with significantly polluted air. The locations of the tropopause in NCEP, GEOS-Chem and GEM-AQ are overlaid as black curves.

• MLS shows that for over 10% of the days in spring, air with significant Asian pollution reaches to North America in the upper troposphere.
• The observed probability is slightly smaller at the Northeast Pacific, even though it is larger over Asia, in summer than in spring.
• GEOS-Chem generally show smaller occurrence probabilities than MLS, except at middle and high latitudes in spring.
• GEM-AQ show comparable occurrence probabilities to MLS in summer. However, the probabilities in the model results are larger than MLS in winter and fall and smaller than MLS in spring.

3 Regional monthly mean CO and its surface sources

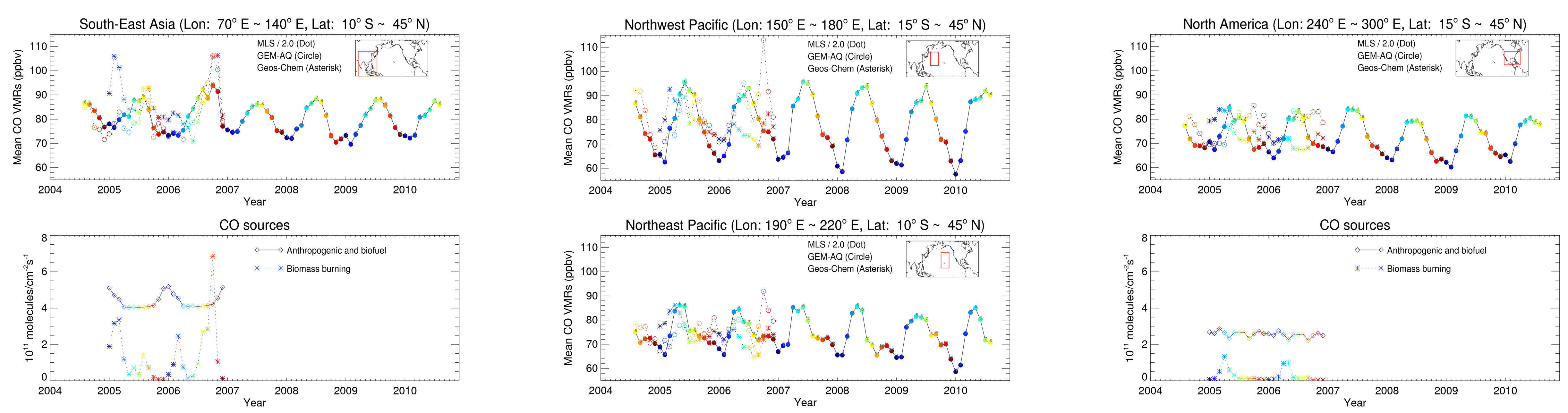
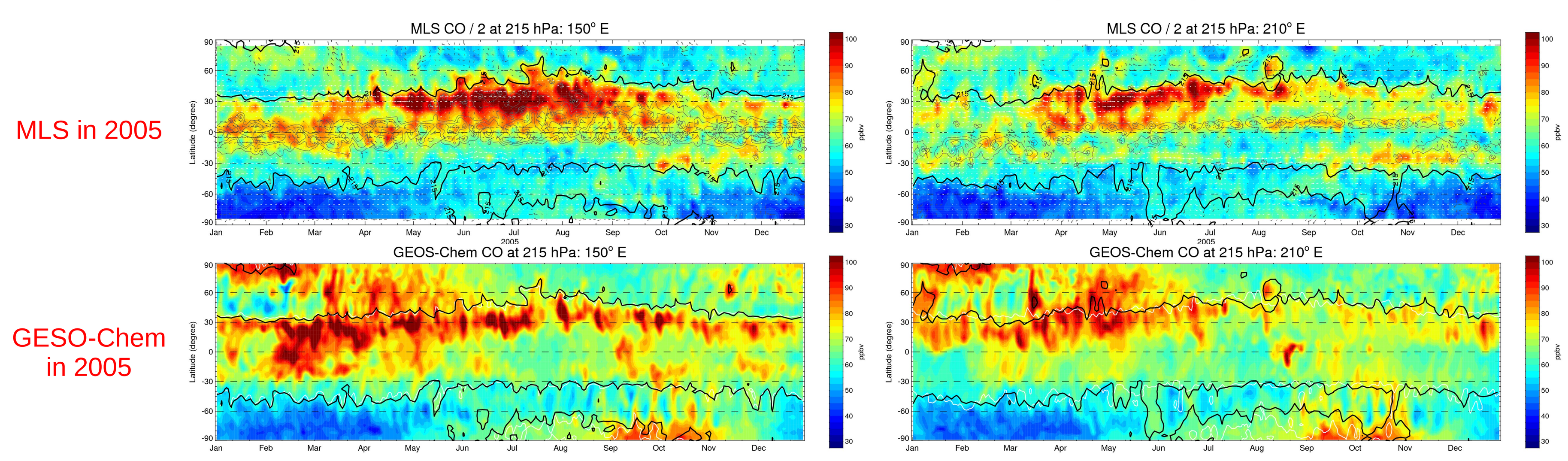


Figure 5 Monthly means of measured and simulated CO over (Top left panel) South-East Asia, (Top middle panel) the Northwest Pacific, (Bottom middle panel) the Northeast Pacific, and (Top right panel) North America. Monthly means of surface CO sources over South-East Asia and North America are plotted in the bottom-left and bottom-right panels, respectively. Different colors represent different months (also labeled with numbers atop the dots).

• No long-term trend of CO abundance is evident in MLS observations in the upper troposphere.
• GEOS-Chem shows larger CO abundance than MLS and GEM-AQ over South-East Asia in Jan-Feb-March 2005.
• GEM-AQ shows larger CO abundance than MLS and GEOS-Chem over the North Pacific in Oct 2006.
• GEM-AQ over-estimates CO in fall and GEOS-Chem has smaller monthly variabilities than MLS over North America.

4 Time-latitude cross-sections over the West Pacific (150° E) and the East Pacific (210° E) in years 2005 and 2006



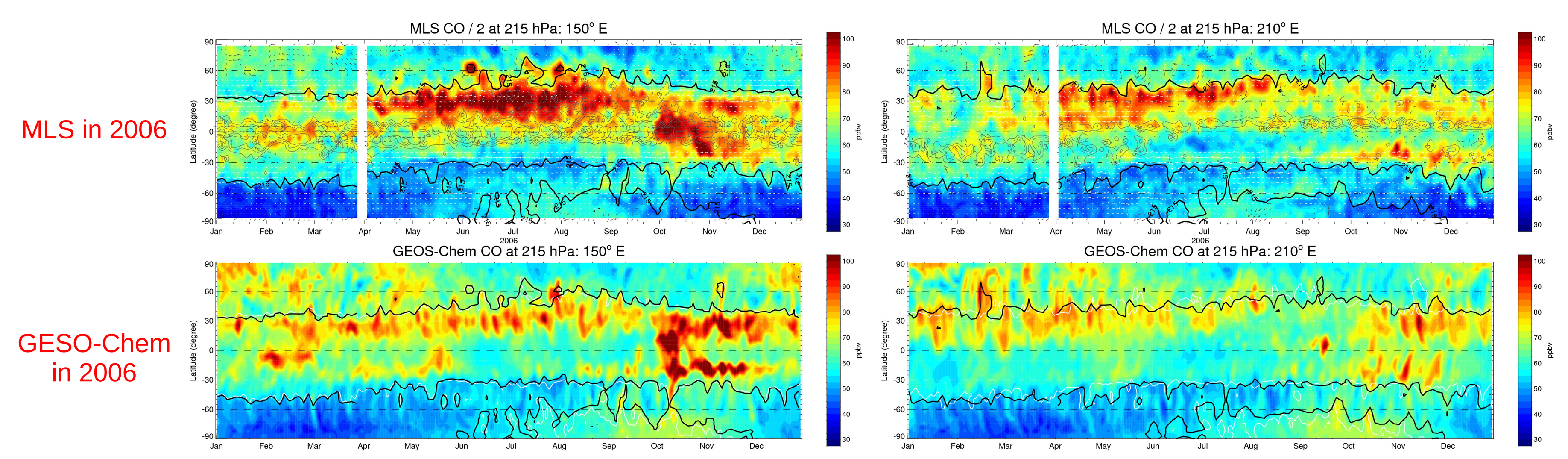


Figure 6 Time-latitude cross-sections of MLS CO measurements and GEOS-Chem simulation over the West Pacific and the East Pacific in year 2005. Grey contours in the top panels are for MLS IWC (mg/m^3). White and black vectors in the top panels show the NCEP westerly and easterly winds, respectively. Black and white curves are the locations of tropopause at 215 hPa in NCEP and GEOS-Chem, respectively.
• MLS observations show large CO abundances are transported along the northern westerly jet south of the location of the tropopause in Apr-Aug, and show little transport in the lower stratosphere. As a result, large meridional CO gradients are seen at the northern tropopause during the period.
• GEOS-Chem reproduces the transport of large CO abundances along the northern westerly jet in summer, but shows larger CO in the northern lower stratosphere in spring.

Figure 7 The same as Figure 6, but for year 2006.

• MLS observations show strong Asian outflow and trans-Pacific transport in late spring and summer in year 2006 as in year 2005, as well as the large CO abundance in subtropics in Oct-Nov 2006 after the big Indonesia fire (Nassar et al. 2009).
• GEOS-Chem simulations are significantly smaller than MLS measurements in late spring and summer. However, it reproduces the transport of equatorial fire emission in subtropics in fall season.

5 Summary

• MLS observations show strongest Asian upper tropospheric outflow of air pollution during summer, but more air with large CO abundance is transported to North America in spring than in the other seasons. For over 10% of the days in spring, air with CO over 90 ppbv reaches to North America.
• GEOS-Chem and GEM-AQ can reproduce the long range transport of air pollution in the upper troposphere. However, GEOS-Chem shows significantly weaker Asian outflow and trans-Pacific transport in summer, whereas GEM-AQ shows stronger transport in winter and fall than MLS. GEOS-Chem generally shows smaller standard deviations of CO abundance in the transport plume over the North Pacific within each season while GEM-AQ generally shows larger standard deviations than MLS.
• The monthly mean series do not show an evident changing trend of CO abundance in the upper troposphere. Comparisons on regional monthly mean CO show GEOS-Chem produces large CO abundances over South-East Asia in Jan-Mar 2005 and GEM-AQ produce large CO over Asia and the North Pacific in Oct 2006.
• GEOS-Chem shows larger CO transport in the lower stratosphere in spring 2005 while MLS shows little transport of CO in the lower stratosphere. Both MLS and GEOS-Chem demonstrate the outflow of equatorial biomass burning emissions in subtropics in fall 2006.

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